Fe/MgO/FeCo(100) Epitaxial Magnetic Tunnel Junctions Prepared by Using In situ Plasma Oxidation

著者	高梨　弘毅
journal or publication title	Journal of Applied Physics
volume	93
number	10
page range	8041-8043
year	2003
URL	http://hdl.handle.net/10097/47276

doi: 10.1063/1.1557338

Fe/MgO/FeCo(100) epitaxial magnetic tunnel junctions prepared by using *in situ* plasma oxidation

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(Presented on 14 November 2002)

Fe/MgO/FeCo epitaxial magnetic tunnel junctions (MTJs) were prepared on MgO(100) single crystal substrates by using *in situ* plasma oxidation for the formation of MgO barriers. The epitaxial relationship of Fe(001)/MgO(001)/FeCo(001) and Fe[100]//MgO[110]//FeCo[100] in the junctions was observed by reflection high-energy electron diffraction. Tunneling transport was clearly observed at low temperatures below about 150 K, and the barrier height of MgO is estimated to be 0.9 eV, which is smaller than the value expected from half of the band gap of bulk MgO. Tunnel magnetoresistance of 23% and 20% was observed at 4.2 and 77 K, respectively. The results suggest that plasma oxidation is useful for fabricating epitaxial magnetic tunnel junctions. [DOI: 10.1063/1.1557338]

I. INTRODUCTION

Epitaxial magnetic tunnel junctions (MTJs) have opened new perspectives in spin-dependent tunneling transport.[1–8] For example, large tunnel magnetoresistance (TMR) was observed in epitaxial MTJs, e.g., Fe/MgO/FeCo(100)[4] and $Ga_{1-x}Mn_xAs/AlAs/Ga_{1-x}Mn_xAs(100)$.[5] The large TMR effects are supported by theoretical calculations, and huge TMR (~1000%) is predicted in an ideal Fe/MgO/Fe(100) MTJ.[9,10] Epitaxial MTJs also have an advantage in controlling structures of a nanometer scale. Quantum well effects on TMR are successfully observed in MTJs with a few atomic layer thick nonmagnetic layer epitaxially grown on the bottom ferromagnetic electrode.[6,7] Controlling the insulator thickness and roughness brings about an antiferromagnetic coupling in Fe/MgO/Fe/Co MTJs.[11]

MgO is one of the most attractive materials suitable for an epitaxial insulating barrier because of the small lattice mismatch for Fe (3.7%) and the large TMR.[4,9,10] Compared to amorphous Al–O barriers, however, the preparation process of epitaxial MgO barriers has been little investigated. Owing to the difficulty in formation of epitaxial MgO barriers, very few experimental results have been reported to date on the preparation and TMR of epitaxial MTJs consisting of Fe (or Fe alloy) electrodes and a MgO barrier.[4,8,11,12] To our knowledge, sizable TMR has been obtained only by two research groups in the Fe/MgO/FeCo(100) MTJs prepared by using laser ablation and molecular beam epitaxy.[4,8] In this study, we have attempted to prepare Fe/MgO/FeCo epitaxial MTJs through the use of conventional ultrahigh vacuum deposition techniques and *in situ* plasma oxidation, and have characterized magnetic and transport properties of the MTJ samples.

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II. EXPERIMENTAL PROCEDURE

Fe/MgO/FeCo MTJs were prepared on MgO(001) single crystal substrates with molecular beam epitaxy equipment (Eiko EB-5K). A flat Fe bottom electrode of 200 Å in thickness was formed on a MgO(001) substrate by electron beam deposition of Fe at room temperature (RT) and postannealing at 200 °C. The surface roughness evaluated by atomic force microscopy (observation area: $1\times1\ \mu m^2$) was ~5 nm in peak-to-peak roughness, which is comparable to that of MgO substrates. A 8 Å thick Mg layer was deposited on the Fe bottom electrode at RT, and was subsequently oxidized in oxygen-argon plasma for 1 to 2 min, where the oxygen-argon plasma was generated by applying 10 W rf power to 4 Pa Ar-20%O_2 gas. Since the number density of Mg atoms in MgO is 25% larger than that in pure metal Mg, the nominal thickness of a MgO layer made from an 8 Å Mg layer is considered to be 6.4 Å (=8/1.25). 19 and 26 Å thick MgO layers were prepared by three and four times repetition of this process. A 200 Å thick $Fe_{50}Co_{50}$ layer as a top electrode was deposited on the MgO barrier. When the samples for transport measurements were prepared, a couple of shadow masks were used for the cross-pattern of MTJs. Changing shadow masks was performed without exposing sample surfaces to air. The junction area was $0.5\times0.5\ mm^2$.

Crystal structures and epitaxial growth were monitored by *in situ* reflection high-energy electron diffraction (RHEED). Magnetic properties were characterized with a superconducting quantum interference device (SQUID) magnetometer. Electrical resistance and TMR were measured by a conventional dc four-probe method.

III. RESULTS AND DISCUSSION

Figure 1 shows RHEED patterns for Fe, MgO, and FeCo surfaces in a MgO[100] azimuth in a MgO(001)/Fe(200 Å)/MgO(19 Å)/FeCo(200 Å) junction. The RHEED pattern for the Fe bottom electrode indicates that the Fe surface is relatively flat and a non-1×1 surface structure appears, which is

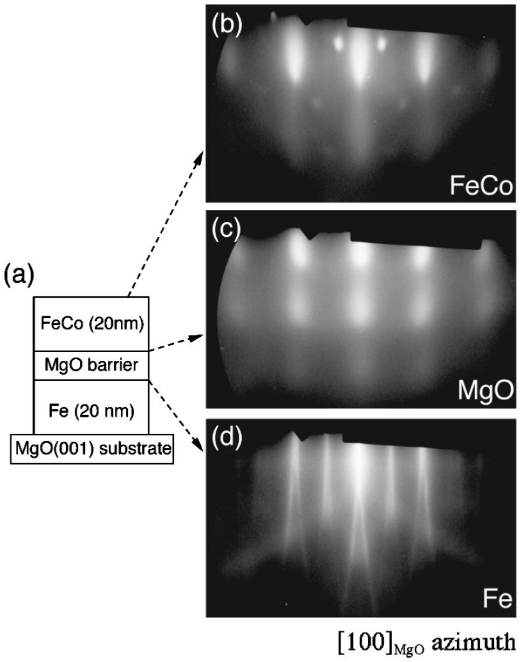

FIG. 1. (a) Schematic illustration of a MTJ structure, and RHEED patterns for (b) FeCo, (c) MgO, and (d) Fe surfaces in a MgO(001)/Fe(200 Å)/MgO(19 Å)/FeCo(200 Å) junction. The incident electron beam is along MgO[100].

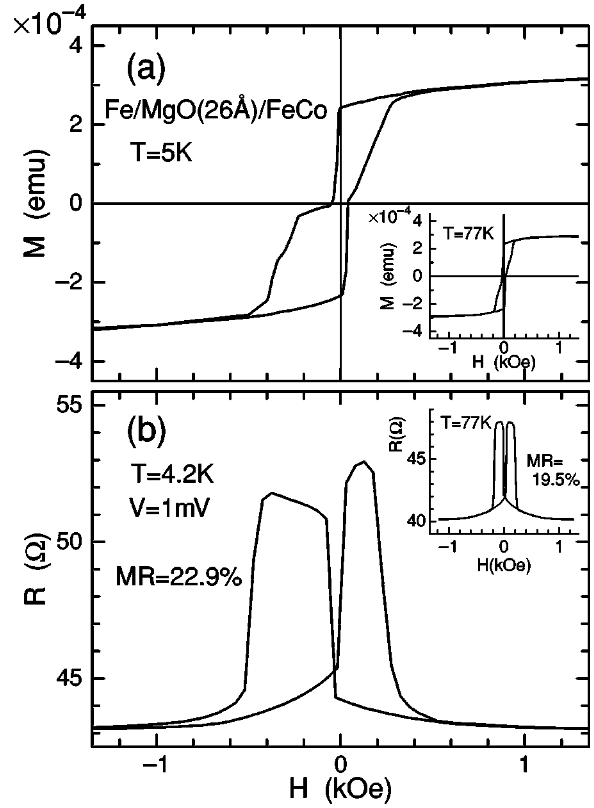

FIG. 3. (a) Magnetization curve at $T=5$ K and (b) TMR at $T=4.2$ K for a MgO(001)/Fe(200 Å)/MgO(26 Å)/FeCo(200 Å) junction. The insets show data at $T=77$ K.

often seen for Fe(001) layers grown epitaxially on MgO(001).[13] The RHEED pattern shown in Fig. 1(c) exhibits the formation of an epitaxial MgO barrier onto the Fe(001) bottom electrode. This suggests that the repetition of thin Mg layer deposition and subsequent plasma oxidation is a useful process to form epitaxial MgO barriers in MTJs. Epitaxial growth of the FeCo top electrode is also confirmed by the RHEED pattern [Fig. 1(b)]. A few extra spots in Fig. 1(b) are probably due to reflection from facet planes of the three-dimensionally grown FeCo layer. The epitaxial relationship of Fe(001)/MgO(001)/FeCo(001) and Fe[100]//MgO[110]//FeCo[100] was confirmed from the RHEED observation by rotating the MTJ sample.

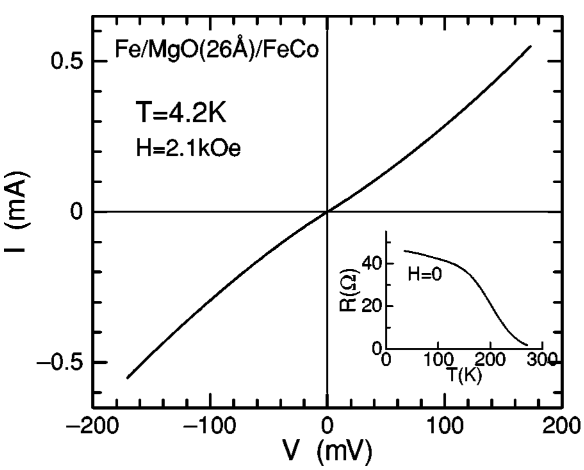

FIG. 2. $I-V$ characteristics at $T=4.2$ K and $H=2.1$ kOe in a MgO(001)/Fe(200 Å)/MgO(26 Å)/FeCo(200 Å) junction. The inset shows temperature dependence of tunnel resistance at $H=0$ Oe in the same sample.

Current–voltage ($I-V$) characteristics at 4.2 K for a MgO/Fe(200 Å)/MgO(26 Å)/FeCo(200 Å) junction is shown in Fig. 2. The nonlinear $I-V$ curve suggests that the electrical current is due to electron tunneling between the ferromagnetic electrodes through the MgO barrier. The barrier width and height estimated from the numerical fitting of Simmons' formula[14] to the experimental result are 22 Å and 0.9 eV, respectively. The estimated barrier width of 22 Å is a little smaller than the nominal thickness of the MgO barrier (26 Å). This is interpreted by the fact that the MgO barrier is not perfectly flat and most of the tunnel current flows through the area where the MgO barrier is relatively thin. The estimated barrier height is high enough to observe the TMR effect but is much smaller than the value expected from half of the band gap of bulk MgO. The barrier height of 0.9 eV is comparable to the value reported for a Fe/MgO/FeCo(001) MTJ with a MgO barrier deposited by laser ablation (1.1 eV).[3] The tunneling transport through the MgO barrier is supported by the negative temperature coefficient of resistance below ~150 K shown in Fig. 2. However, above ~150 K the resistance decreases dramatically, and the exponential temperature dependence of resistance above ~150 K is interpreted in terms of a thermal activation process of conduction electrons. A possible origin of the semiconducting current channel that opens above ~150 K is considered to be hopping conduction through defects in the MgO barrier. In order to observe tunneling transport at room temperature, it is necessary to remove the additional current channels by improving the quality of MgO barriers.

Figure 3(a) and 3(b) show magnetization curves and TMR at $V=1$ mV for a MgO(001)/Fe(200 Å)/MgO(26 Å)/FeCo(200 Å) junction, respectively. It is seen that the resistance changes corresponding to the magnetization curve, although the switching field and the saturation field etc. are a little different between the samples for magnetization and transport measurements. Asymmetric field dependence probably originates from exchange magnetic anisotropy from an antiferromagnetic oxide layer on the surface of the top electrode because no protective layer is deposited. Antiparallel alignment of the magnetizations of Fe and FeCo electrodes is realized, and TMR of 23% is successfully observed at 4.2 K for the MTJ. This value of TMR is smaller than that reported for the Fe/MgO/FeCo MTJ prepared by using laser ablation,[3] suggesting that TMR in the MgO based epitaxial MTJs depends on the barrier and interface structures as well as alumina-based MTJs. With increasing temperature up to 77 K, asymmetry in the field dependence of magnetization and TMR disappears, and TMR of about 20% is still observed. Similar magnetic and transport properties were observed for a MgO(001)/Fe(200 Å)/MgO(19 Å)/FeCo(200 Å) junction. TMR depends on the thickness of the MgO barrier, resulting in TMR of 14% at $T=4.2$ K and $V=1$ mV. Further experiments on structural analyses and optimization of the plasma oxidation process are in progress to achieve larger TMR effects.

IV. CONCLUSION

Fe/MgO/FeCo epitaxial magnetic tunnel junctions (MTJs) were prepared on MgO(001) single crystal substrates. Deposition of 8 Å thick Mg and subsequent plasma oxidation were repeated for the formation of MgO barriers on Fe(001) bottom electrodes. The epitaxial relationship of Fe(001)/MgO(001)/FeCo(001) and Fe[100]//MgO[110]//FeCo[100] in the junctions was confirmed by RHEED observation. Nonlinear I–V characteristics and negative temperature coefficient of resistance were clearly observed at low temperatures below about 150 K, showing tunneling transport in the Fe/MgO/FeCo MTJs. The barrier height of MgO is estimated to be 0.9 eV, which is smaller than the value expected from half of the band gap of bulk MgO. TMR of 23% and 20% was successfully observed at 4.2 and 77 K, respectively. The results suggest that plasma oxidation is useful for fabricating epitaxial magnetic tunnel junctions.

ACKNOWLEDGMENT

This work was partly supported by the IT-Program of RR2002 from MEXT.

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